# SPECTRAL EXTINCTION COEFFICIENTS OF SOOT AGGREGATES FROM TURBULENT DIFFUSION FLAMES[1]

by

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## Introduction

Accurate knowledge of the spectral variation of the refractive indices of soot is necessary in order to estimate continuum radiation from luminous flames and to develop *in-situ* optical techniques for measuring soot properties. However, there are fairly large variations among the soot refractive indices reported in the literature, implying uncertainties about which values should be used in a particular application, see, for example, Tien and Lee[1], Chang and Charalampopoulos[2], and references cited therein for a complete discussion. Moreover, results regarding the effects of fuel type (H/C ratio) on soot refractive indices are also contradictory. Early work of Dalzell and Sarofim[3], and Lee and Tien[4] suggested that soot refractive indices were relatively insensitive to fuel type. On the other hand, later work of Habib and Vervisch[5], and Charalampopoulos *et al.*[6] indicated significant effects of fuel type on soot refractive indices. However, Sivathanu *et al.*[7] recently reported that their measurements were most consistent with the values reported by Dalzell and Sarofim[3], while finding only a weak dependence of fuel type on soot refractive indices. In view of these observations, the main objective of the present study was to evaluate the capabilities of the soot refractive indices reported in the literature to treat the spectral extinction properties of soot aggregates, and to investigate the effect of fuel type on refractive indices.

The present measurements included the extinction coefficients of soot in the wavelength region of 0.2-5.2 μm. Four hydrocarbon fuels were considered to study the effect of fuel type (H/C ratio) on spectral refractive indices of soot. Test conditions were limited to the fuel-lean (overfire) region of buoyant turbulent diffusion flames in the long residence time regime, where soot morphology, and soot scattering properties at 0.514 μm, were known from earlier work [8,9]. The mean spectral radiative properties were predicted based on Rayleigh-Debye-Gans theory for polydisperse fractal aggregates (RDG/PFA) because this approach has been found to be the best approximation to represent the optical properties of soot aggregates [9-13]. The results of the present spectral extinction measurements were combined with earlier findings in order to evaluate existing refractive indices in the literature. The spectral soot refractive indices reported by Chang and Charalampopoulos[2], Dalzell and Sarofim[3], and Lee and Tien[4], were chosen for comparison with the present measurements because they are frequently used in heat transfer and combustion studies and they also involve different experimental and data inversion methods. The following discussion of the study is brief; see Köylü and Faeth[14] for details.

## Experimental Methods

The present test arrangement was the same as for the earlier light scattering measurements of Köylü and Faeth[9]. The apparatus consisted of a 50 mm diameter water-cooled burner which provided buoyant turbulent diffusion flames in the long residence time regime. Four gaseous hydrocarbon fuels - acetylene, propylene, ethylene and propane - were used to vary H/C ratios in the range 0.08 to 0.22.

The spectral extinction coefficients of overfire soot were measured in the wavelength region of 0.2-5.2 μm. Three different light sources - a 30 W deuterium lamp, a 100 W QTH lamp and a 100 W IR element - were used in the wavelength regions 0.20-0.35, 0.35-2.0 and 2.0-5.2 μm, respectively. $CaF_2$ lenses were used for collimating/focusing the light due to the wide range of wavelengths considered. The light was modulated by an enclosed chopper. The light intensities were measured by a pyroelectric detector with a flat response in the wavelength range of 0.2-40 μm. Different interference filters were used in front of the detector to make measurements at the wavelength of interest. The strong gas absorption bands of $H_2O$ at 2.7 μm, and $CO_2$ at 4.3 μm were avoided during these spectral measurements.

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[1]Prepared for the Fall Meeting of the Eastern States Section of the Combustion Institute, Worcester, MA, 16-18 October 95.

## Results and Discussion

Scattering at 0.514 μm. Measured phase function from the earlier scattering measurements of Köylü and Faeth[9] at 0.514 μm, along with the predictions from the various approximate theories, are plotted as a function of scattering angle in Fig. 1 for acetylene soot. The predictions of the RDG/PFA theory are in good agreement with measurements whereas neither Mie theory for an equivalent diameter nor the Rayleigh approximation can predict the angular trends of phase function. These results suggest the importance of aggregate scattering and justifies the use of RDG/PFA approximation to estimate spectral properties of soot aggregates. Additionally, these measurements implies F(m)/E(m) = 0.78 with a standard deviation of 0.09 over all the fuels, where F(m) and E(m) are functions of the refractive index [9]. Since a value of E(m) = 0.23 seems to be reasonable [2-4], this yields a complex refractive index of m = 1.54 + 0.48i at 0.514 μm, which is in reasonably good agreement with the measurements of Chang and Charalampopoulos[2], and Dalzell and Sarofim[3]. However, the refractive indices at this wavelength given by Lee and Tien[4] yield F(m)/E(m) = 1.55, which is almost 100% higher than the measured value.

Spectral Extinction Properties of Soot. Measurements of optical thickness in the wavelength range 0.2-5.2 μm, are illustrated in Fig. 2 for all the fuels. The results yielded a maximum extinction coefficient in the ultraviolet region at a wavelength of approximately 0.25 μm for all fuels. Menna and D'Alessio[15] and Chang and Charalampopoulos[2] reported similar observations, which seems to be caused by a resonance of soot refractive indices at this wavelength range. In contrast, the behavior of spectral extinction coefficients in the infrared is rather monotonic, as expected.

Measured and predicted specific extinction coefficients are plotted as a function of wavelength in Fig. 3. The spectral measurements are for the overfire soot aggregates emitted from a propane/air flame, while the predictions are based on RDG/PFA theory using the refractive indices of Dalzell and Sarofim[3], Lee and Tien[4], and Chang and Charalampopoulos[2]. The refractive indices from the three studies overestimate the observed extinction levels by up to a factor of three in the ultraviolet. However, the values from Chang and Charalampopoulos[2] correctly predict the properties of the resonance of refractive indices near 0.25 μm. Although the measurements of Dalzell and Sarofim[3], and Lee and Tien[4] do not extend into this short wavelength region, their dispersion models fail to indicate the presence of the resonance phenomena, as quantified by Menna and D'Alessio[15]. Thus, the magnitude as well as the trend of the refractive indices given by Refs. [3,4] do not agree with present measurements in the UV.

The refractive indices from the three sources yield predictions that start deviating from measurements as the wavelength increases from visible into the infrared. Although the values from Dalzell and Sarofim[3] yield best agreement with measurements in this wavelength range, all three data sets underestimate the measurements of specific extinction coefficients. This is probably due to their low E(m) values in the infrared, as will be demonstrated in the following by taking the ratio of E(m) functions at wavelengths of $\lambda_2$ = 5.2 μm and $\lambda_1$ = 0.514 μm [14]:

$$E(\lambda_2)/E(\lambda_1) = (K_e(\lambda_2)/\ K_e(\lambda_1))\ (\lambda_2/\lambda_1)\ (1+\rho_{sa}(\lambda_1)) \qquad (1)$$

where $K_e$ and $\rho_{sa}$ are the extinction coefficient and the ratio of the total scattering and absorption coefficients, respectively. Noting that all the quantities on the right-hand side are known from the spectral measurements, Eq. (1) yields E(5.2 μm)/E(0.514 μm) = 1.89 with a standard deviation of 0.18 for all the fuels. This ratio is given by Dalzell and Sarofim[3], Lee and Tien[4], and Chang and Charalampopoulos[2] as 1.38, 1.00, and 1.21, respectively. Thus, the closest agreement is obtained from the refractive indices of Dalzell and Sarofim[3], underestimating the measured value by 27%. Nevertheless, the differences between predictions and measurements are typically 50-100% at long wavelengths.

The emissivities of the soot aggregates are illustrated as a function of wavelength for propylene in Fig. 4. Spectral emissivities are obtained using the measurements of specific extinction coefficients. Similar to the results for specific extinction coefficients, the predicted emissivities are overestimated in the UV and underestimated in the IR. On the other hand, measurements and predictions are in good agreement for visible wavelengths. Among the three, the refractive indices from Dalzell and Sarofim[3] appear to be yielding best estimates of emissivities, followed by the results of Chang and Charalampopoulos[2]. It should be mentioned that the results for other fuels were similar.

Experimental Correlation for Spectral Properties. Several authors (see, for example, Siegel and Howell[16], and Siddall and McGrath[17]) have reported that the specific extinction coefficient of soot, defined as the extinction coefficient per unit volume fraction, varied with wavelength according to:

$$K_e/f_v \propto \lambda^{-\alpha} \qquad (2)$$

The use of Eq. (2) is practical in radiative heat transfer calculations for soot, since the expression does not involve the complications of refractive indices. Thus, a similar correlation was sought for the present test conditions. The least-square fits of the measurements in the wavelength range of 0.514-5.2 μm yielded a value of $\alpha = 0.83$ with a standard deviation of 0.04 for the four fuels considered in this study. The values suggested by the refractive indices of Refs. [2-4] are in the range of 0.94-1.21, which are 13-47% higher than the present measurements. Additionally, the above experimental correlation implies that E(m) should slightly increase with increasing wavelength, *i.e.*, $E(m) \propto \lambda^{0.17}$ in the infrared. This also suggests that the refractive indices of soot are weak functions of wavelength in this part of the spectrum. This behavior, however, differs from the behavior of avaliable refractive indices in the infrared, which either gives a decrease [3,4], or almost no variation [2] of E(m) with increasing wavelength in the infrared. These overall deviations of predictions from measurements may result in significant differences (20-40%) in the total emittance for typical flame temperatures.

Effect of Fuel Type on Soot Refractive Indices. The present spectral extinction measurements, together with previous scattering results, showed that the ratios of F(m)/E(m) at 0.514 μm, the ratios of values of E(m) at 5.2 μm and 0.514 μm, and the values of α, representing the spectral variation of refractive indices in the range of 0.514-5.2 μm did not change with fuel type within experimental uncertainties. All three measurements suggest that soot refractive indices are relatively independent of fuel type for the visible and infrared spectral ranges for the present test conditions. In view of similar observations of Sivathanu *et al.*[7] for soot within laminar premixed flames, this weak dependence may also apply to other flame environments, simplifying both *in-situ* soot volume fraction determinations and radiative heat transfer calculations. However, additional studies in various flames will be required in order to firmly establish the insensivity of refractive indices to fuel type.

**Acknowledgements.** This research was sponsored by the Building and Fire Research Laboratory of the National Institute of Standards and Technology, Grant No. 60NANB1D1175, and by NASA, Grant No. NAG3-1245. One of the authors (Ü. Ö. K.) would like to acknowledge the partial support by AFOSR, Grant No. 94-0143.

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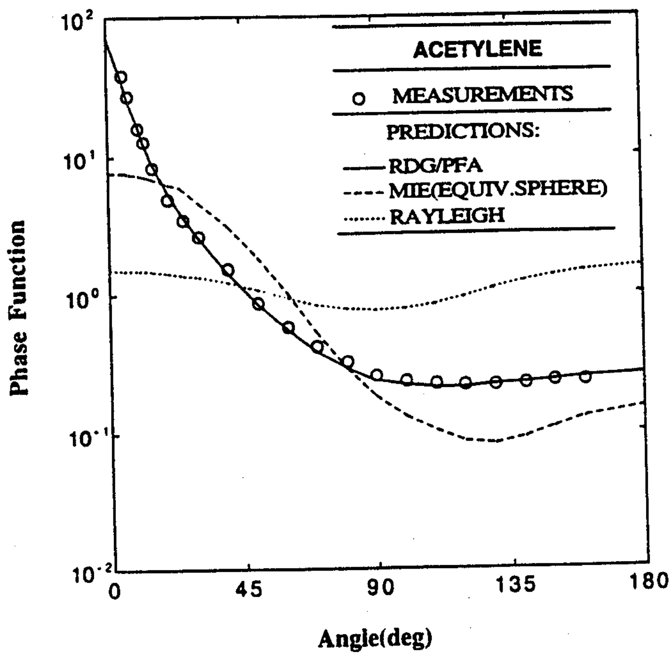


Figure 1. Various theoretical predictions and measurements of phase function of overfire soot from turbulent acetylene/air diffusion flame.

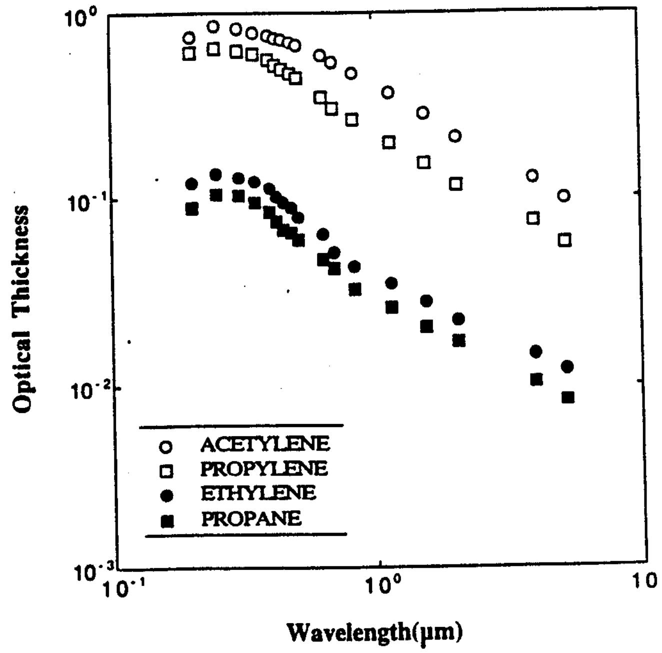


Figure 2. Measurements of optical thickness as a function of wavelength for overfire soot from turbulent hydrocarbon diffusion flames.

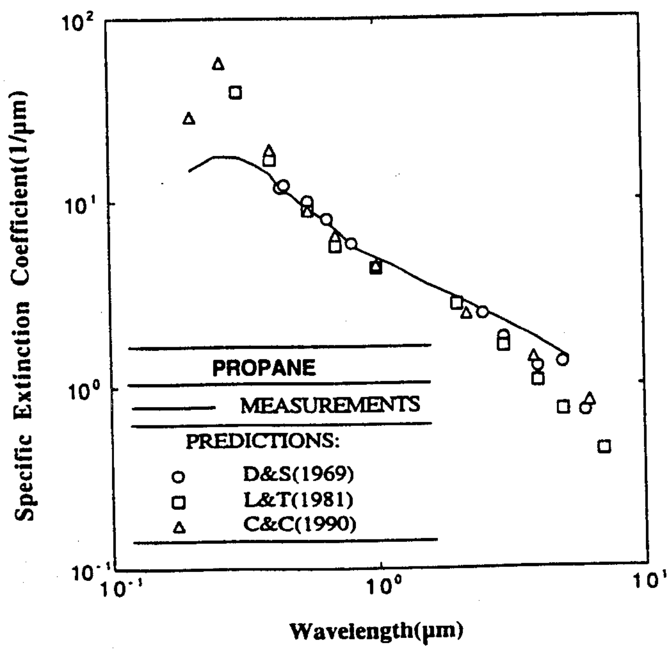


Figure 3. Predictions of various refractive indices and measurements of specific extinction coefficient of overfire soot from turbulent propane/air flame.

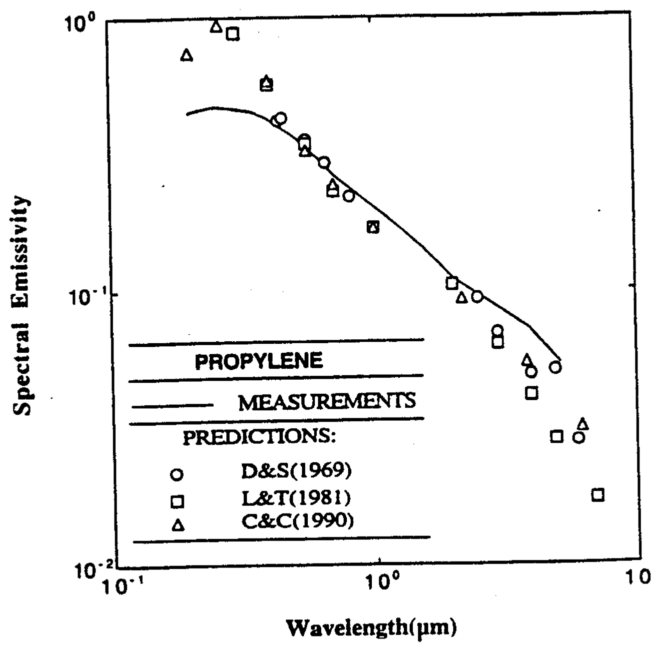


Figure 4. Predictions of various refractive indices and measurements of emissivity of overfire soot from turbulent propylene/air diffusion flame.